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CdTe/ZnTe quantum dots—growth and optical properties

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Abstract

This paper gives an overview of molecular beam epitaxy growth aspects and of optical properties of CdTe quantum dots grown on ZnTe by self-assembly. It is shown that quantum dots in this material system can be obtained either by depositing CdTe at a high substrate temperature or by subjecting CdTe layer to a healing process, up to 70 s long before its capping or, eventually, by applying these two methods simultaneously. Moreover it is found, that one can also use the atomic layer epitaxy method to achieve the formation. From optical measurements performed on large quantum dot ensembles it is found out that the quantum dot emission is much broader than that from quantum wells, and that it is observable up to much higher temperatures, which indicates strong exciton localization. The latter is also evidenced by an insensitivity of the decay time of the exciton recombination (~300 ps) to the temperature. From the presence of a second, very long decay time (~5 ns) and from disappearance of the sharp lines related to recombination in single dots, the acoustic phonon scattering of excitons is found to play an important role in these quantum dot structures. From a magnetic field dependence of the single dot emission energy, the exciton effective g-factor and spatial extension of the exciton wave function are deduced to be equal to -3 and 3 nm, respectively. Both the g-factor and the value of the diamagnetic shift are found to be independent of the energy of the quantum dot emission at $B=0$ T and of the in-plane symmetry of its potential.

Keywords: Cadmium telluride; Molecular beam epitaxy; Nanostructures; Optical spectroscopy

1. Introduction

In this paper several aspects of growth and optical properties of self-assembled CdTe quantum dots (QDs) in ZnTe matrix are reviewed. This particular material system is interesting from the point of fabrication of QDs [1–3] since the lattice mismatch between these two constituents is similar to that of InAs/GaAs [4], which is the most widely studied system in the context of QDs. However, as one can expect, different chemical properties of II–VI compounds such as diffusion coefficients could influence the formation process of this type of nanostructure [5].

We indicate that both healing of CdTe layer after its deposition on ZnTe as well as growth of CdTe at the substrate temperature higher than that used typically for obtaining high quality quantum wells (QWs) [6] result in the formation of three-dimensional structures with nanometric scale on a two-dimensional layer. Moreover, the formation of QDs is also observed when a CdTe layer is grown by atomic layer epitaxy, as reported by another group [3].

The formation of the QDs is evidenced by optical spectroscopy measurements. The low temperature luminescence (PL) emission is characterized by a broad band [5] which exhibits roughly temperature independent decay time [7–9] equal to 300 ps, as long as the effects of carrier redistribution between the dots do not play an important role [8,10]. When decreasing the size of the excitation laser spot this inhomogeneously broadened emission splits into a series of sharp lines with the linewidth of approximately 100 μeV [11–13]. However, with an increase of the temperature the sharp line pattern disappears, as does the intensity of the emission, which is characterized by a very long decay time of the order of several nanoseconds [14–17]. These effects are ascribed to a broadening of the single dot PL line caused by acoustic phonon scattering of excitons [14,17].

The results of PL experiments performed with ultrahigh spatial and spectral resolution on CdTe QDs show that the optical properties of an exciton localized by a single QD potential strongly depend on the in-plane symmetry of the particular QD. Namely, for elongated QD the exciton line splits into linearly polarized doublet [9,13,18]. Furthermore, it is found that the reduction of the dimensionality results in an enhancement of the

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effective excitonic g-factor. Surprisingly, the value of g-factor is found to be independent either of the QD confinement energy as well as of the particular in-plane symmetry of the dot. From the magnetic field dependence of the emission energy the value of exciton diamagnetic shift is obtained [11] from which, in turn, the extension of excitonic wavefunction is evaluated to be equal to 3 nm. This value agrees reasonably with the average size of the QDs estimated from transmission electron microscopy data [1].

The present paper is organized as follows. After a detailed description of the samples which includes not only the growth conditions but also the procedure of obtaining small apertures in a metal mask (making experiments with single QDs possible), the experimental set-ups which were used for investigating the optical properties of QDs are briefly presented. In the next part the results of optical experiments are discussed starting from PL characterization of the samples. Then the temperature and the time dependencies of the PL emission are described. The experimental part is completed by a description of microluminescence spectroscopy as a function of the temperature and the magnetic field.

2. Samples and experimental details

The investigated structures were grown by molecular beam epitaxy on (100)-oriented GaAs substrates. In order to avoid dislocations formed due to lattice mismatch between GaAs and CdTe, first buffer layers of CdTe and ZnTe with the thickness ranging between 4 and 5 μm were grown. Since one of the purposes of viewed studies was to describe the influence of the growth conditions on formation of the quantum dots, several procedures of CdTe deposition were employed. Usually, the growth of high quality CdTe quantum wells (QWs) was carried out at a relatively low substrate temperature, namely at approximately $T=300$ °C [6]. At such temperatures the surface diffusion is quite low which results in sharp interfaces between barriers and the well layers. From that point of view an increase of the substrate temperature enhances the mobility of atoms on the surface which could lead to the formation of structures which can localize excitons stronger than in QWs. Another parameter that can help in a process of a QD formation could be the time between stopping the growth of the CdTe layer and capping it next with a ZnTe barrier. If that time is long enough one can expect that the atoms can migrate more easily on the surface. Thus, the effect of such a growth interruption is qualitatively similar to an increase of the substrate temperature. However, in the latter approach some amount of the deposited material could be desorbed from the surface.

The results presented in this paper were basically obtained for four groups of structures. All samples are listed in Table 1 together with the substrate temperature during the growth, time of growth interruption (healing), number of CdTe layers as well as the nominal thickness of the CdTe layer. The last two samples containing just a single CdTe layer were grown by the atomic layer epitaxy method. In all these samples the ZnTe capping barrier layers were thick enough to prevent any interaction between neighboring layers of quantum dots [2]. The reason for growing 10 layers of CdTe QDs was to enhance the optical response from each sample. On the other hand, the samples that contain only one layer of CdTe QDs were explored by means of single dot spectroscopy methods.

Table 1
Parameters of the samples studied in this paper: substrate temperature, time of healing, nominal CdTe thickness and the number of CdTe layers

Sample number	Substrate temperature (°C)	Healing time (s)	Nominal CdTe thickness (ML)	Number of CdTe layers
1	480	0	5	10
2	300	0	5	10
3	300	70	4	10
4	300	0	4	10
5	420	10	1.5	10
6	420	10	2	10
7	420	10	2.5	10
8	320	0	2	1 (ALE)
9	320	0	4	1 (ALE)

Various experimental methods were employed to investigate the optical properties of these structures. The continuous wave PL was excited non-resonantly above the ZnTe barrier gap energy ($\lambda=457$ nm line of an argon laser). The emission was analyzed by a Jobin-Yvon spectrograph equipped with a LN_2-cooled charge-coupled-device (CCD) detector. The temperature of the sample was changed from 4.2 to 200 K. The exciton dynamics was studied by time-resolved PL utilizing the time-correlated single photon counting technique. In this case the signal, after being dispersed by a monochromator, was detected by a fast microchannel plate. As an excitation source for time-resolved PL experiments the frequency-doubled (370 nm) Ti:sapphire laser giving 6 ps long pulses with 80 MHz repetition was used. The temporal resolution of the set-up was equal to 40 ps.

Apart from these methods, which probe the whole QD ensemble, the techniques that allow studying single quantum dots were also used. It becomes possible basically either by focusing the laser spot on the sample surface or by patterning the sample in order to allow optical probing of a very limited number of dots. Part of the microluminescence measurements was performed in a set-up, which allowed to reduce the spot of the laser down to the 2 μm in a diameter by the microscope objective. The sample was mounted in the continuous-flow helium cryostat and the temperature was changed

from 6 to 100 K. As the excitation the 514 nm line of an argon later was used, while the PL emission was detected by a CCD camera after being dispersed by a double 1 m monochromator. The spectral resolution of the set-up was equal to 20 μeV. However, due to the large surface density of the dots in the studied structures, by the use of the above-described set-up it was not possible to isolate single emission lines even if a sample contained only one layer of the dots. Thus, in order to suppress further the influence of an inhomogeneous broadening of the QD-related PL emission band (arising from the size, chemical composition or strain fluctuations), the opaque metal (Au/Pd) masks with small apertures were fabricated on the sample surface by electron beam lithography and standard lift-off technique. The size of the apertures estimated by atomic force microscopy was found to be as small as 150 nm.

High-resolution PL measurements were carried out at $T=4.2$ K at magnetic fields up to 12 T applied parallel to the growth direction. As the excitation source the 514 nm line of an argon laser was used. The laser beam was focused on the sample surface by using a microscope objective down to a spot with the diameter of approximately 0.7 μm. In order to resolve the polarization of the light, the emission was analyzed either by a linear polarizer or by a quarter wave plate combined with a linear polarizer. For spectral detection the signal was dispersed by 0.64 m Jobin-Yvon monochromator and detected by a LN_2-cooled CCD detector. The spectral resolution of the set-up was approximately 100 μeV.

In the following part the experimental results obtained for the CdTe/ZnTe quantum structures will be described in detail together with the analysis of the data. We will start with the PL characterization of the structures and after that the results of the temperature dependent PL and time-resolved PL experiments will be discussed followed by microluminescence spectroscopy both as a function of temperature and magnetic field.

3. Experimental results and discussion

3.1. Photoluminescence characterization of the structures

Low temperature ($T=4.2$ K) luminescence spectra for all samples listed in Table 1 are presented in Fig. 1. In the case of the sample grown at the high substrate temperature of 480 °C (Fig. 1a) as well as the one subjected to 70 s healing after CdTe deposition (Fig. 1b) substantial differences are visible when comparing the spectra with those obtained for QWs. Namely, in both cases the PL line is broadened and the PL emission shifts towards higher energies.

The most probable explanation of this behavior could be an effective narrowing of the deposited CdTe layer during the growth at such conditions. Namely, at higher

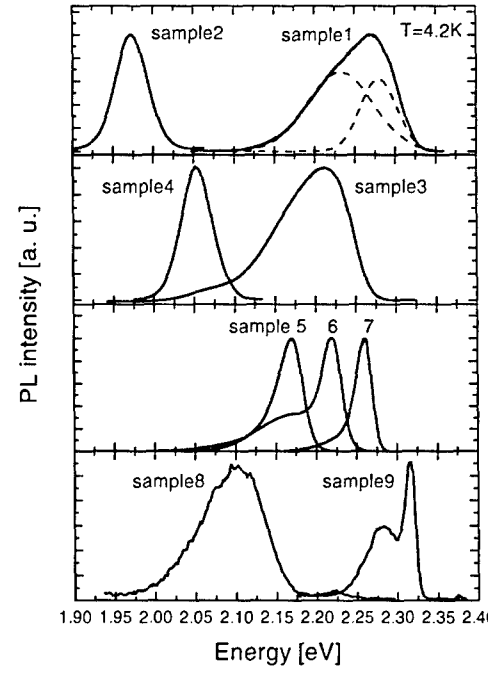

Fig. 1. Low temperature ($T=4.2$ K) PL spectra for all studied structures: (a) growth at high substrate temperature ($T=480$ °C); (b) growth with applying long time healing after CdTe deposition; (c) growth at $T=420$ °C combined with several seconds long healing in Cd atmosphere; (d) growth at 320 °C by atomic layer epitaxy. In two first cases also the spectra of the reference samples are shown.

growth temperatures the desorbtion from the surface becomes very important. This results with the narrowing of the well width. Because the PL emission energy increases with the decrease of the well width one can see by comparing the data displayed in Fig. 1a,b that the narrowing of the well width is much stronger in the case of sample 1. The blue-shift is almost twice as large as in the case of sample subject to the healing. The difference is intuitively understood since at such a high growth temperature the desorbtion processes are expected to be very effective.

Together with the blue-shift and the broadening of the PL lines in these two structures the lineshape of the PL emission becomes more complicated and, in particular, it is no longer possible to approximate it by a single Gaussian. In Fig. 1a the dashed lines show the result of a fitting of two Gaussians to the PL spectra. Both effects, the broadening of the PL linewidth and its complicated shape can not be fully attributed to the evaporation of CdTe during the epitaxy. The broadening is most probably caused by two mechanisms. On the one hand, due to the large lattice-mismatch between CdTe and ZnTe, the CdTe/ZnTe interface is no more ideally flat. Consequently, the well width fluctuations are likely to occur. Then, if the exciton recombination originates from such an ensemble of quantum wells with different thickness, the observed emission should be

obviously much broader than in the case of a quantum well with flat interfaces. On the other hand, apart from QW thickness fluctuations, the interdiffusion of Cd and Zn across the interface seems to be also the process of importance, apparently in the case of sample 1 shown in Fig. 1a.

Both these effects are most probably responsible for the observed broadening of the PL line in the investigated structures. As will be shown in the following, applying either a long healing time or growing the structure at a high temperature lead to a strong excitonic localization. The properties of excitons confined in these two structures are very similar to that observed for self-assembled quantum dots.

In Fig. 1c the PL spectra obtained for the structures where both several seconds long healing (up to 10 s) as well as medium-high substrate temperature ($T=420$ °C) conditions were applied during the growth. In this case for all three structures with a different nominal thickness of CdTe (samples 5, 6 and 7), two emission bands are present in the spectra. On the basis of the results of optical measurements the high-energy emission is assigned to the recombination of excitons in a two-dimensional quantum well, while the low energy line represents the recombination in CdTe quantum dots. Thus, at this stage we may assume that at these growth conditions the self-assembly process takes place, which leads to the formation of quantum dots. From the transmission electron microscopy data reported elsewhere, the average diameter and surface density of QDs in these structures were estimated to be 4 nm and 10^{11}–10^{12} cm^{-1}, respectively [1].

However, as compared with the results reported for InAs QDs in GaAs matrix, the CdTe/ZnTe structures exhibit a substantial different behavior, which indicates that the formation of QDs in the latter system is more complicated than that observed for III-V compounds. Namely, in the III–V systems, when the transformation of the growth mode occurs and the QDs are spontaneously formed on the thin QW called a wetting layer (WL), the thickness of the WL remains unchanged even if the material the dots are formed of is still deposited on the surface [19–21]. In other words, after the transformation all the material is accumulated within the dots: their density as well as their size increases with the increase of the nominal thickness of the QD layer.

Contrary to that, as evidenced in Fig. 1c, with the increase of the nominal thickness of CdTe both the QW and the QD-related emission lines shift towards lower energies. Moreover, together with this shift the ratio between QW and QD recombination intensity is getting smaller. This result indicates that when putting more and more material (i.e. CdTe) on the surface both the two-dimensional QW is getting thicker and the dots are developing in sizes and density, as well.

Such a behavior is even more visible in Fig. 1d, where the PL spectra for the structures grown by atomic layer epitaxy are presented (sample 8 and sample 9). Namely, in the case of the sample with the nominal CdTe thickness of 2 ML one can easily distinguish both QW- and QD-related emissions, while for the sample with the nominal CdTe thickness of 4 ML the QW emission is no longer present. The only visible line is related to excitonic recombination in CdTe quantum dots.

The absence of abrupt growth mode change, from a two-dimensional to three-dimensional one, as observed in CdTe/ZnTe material system, results in a gradual change of reflection high-energy electron diffraction (RHEED) pattern. Contrary to the case of II–VI semiconductor QDs [5], for InAs/GaAs materials sharp transformation of the RHEED pattern, i.e.: from a streaky (characteristic for two-dimensional growth) to spotty (characteristic for three-dimensional growth) was observed [4]. In particular, such a change in RHEED pattern is usually recognized as a signature of QD formation during the growth. On the basis of scanning transmission electron microscopy results obtained for CdSe QDs in ZnSe matrix the following scenario for QD formation was proposed [5]. At the early stage of growth two-dimensional platelets are formed which may be considered as precursors for coherent QDs formation. Eventually, these two-dimensional platelets could form a uniform QW. During this process coherent dots are developed when more and more material the dots are formed of is deposited. The optical spectra shown in Fig. 1 reveal indeed the presence of such two types of structures. This indicates that the QD formation in CdTe/ZnTe system is very similar to that observed in another II–VI material pair, namely CdSe/ZnSe one.

We would like to point out here, that although in order to obtain self-assembled QDs we applied either molecular beam epitaxy (samples shown in Fig. 1c) or atomic layer epitaxy (samples shown in Fig. 1d), the spectral features observed for these two types of structures are qualitatively very similar to each other. One could then conclude that the principle of the dot formation in this system is independent of the growth method applied, although the parameters of obtained QDs such as shape, size or chemical composition can depend on the particular growth procedure.

3.2. Temperature dependence of the photoluminescence

In the following part the influence of the temperature on the PL emission is discussed. From the results of these measurements we can assign the high-energy emission present in the majority of spectra shown in Fig. 1 to a quasi two-dimensional excitonic recombination while the low-energy emission seems to be related to excitons confined in quantum dots. Since, in general,

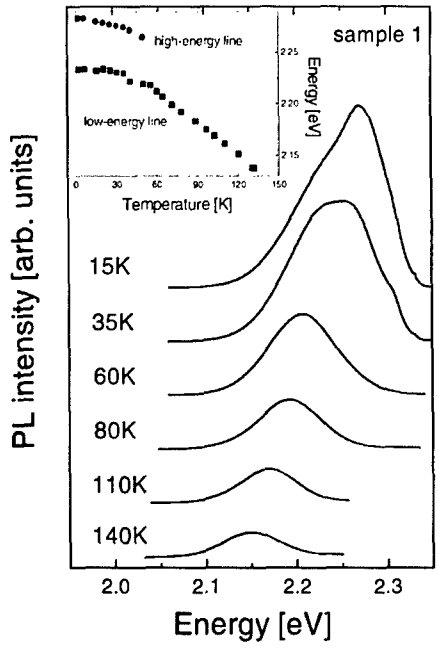

Fig. 2. Temperature evolution of PL emission obtained for sample 1. In the inset the dependence of the emission energy extracted from the spectra is also given.

the observed behavior in the case of all structures was quite similar, we shall consider only the case of the sample 1 (i.e. the one shown in Fig. 1a), which was grown at the substrate temperature equal to 480 °C.

The PL spectra measured for this particular sample at different temperatures are presented in Fig. 2. With an increasing temperature the spectra change substantially. Namely, apart from shifting towards lower energies and decreasing the total intensity of the emission, the shape of the PL line also changes. While the emission observed at $T=4.2$ K shows a rather complicated shape, which requires at least two Gaussians to be used to model the line (Fig. 1a), at sufficiently high temperatures (which is above 100 K) the emission line could be nicely fitted only by a single Gaussian shape. This allows, in turn, to follow the temperature dependence of the energies corresponding to the maximum emission of both bands obtained from the fitting. The result is presented in the inset in Fig. 2. The striking feature is that the observed red-shift of the low energy peak with increasing temperature is much larger than one would expect from the temperature induced shrinking of the CdTe bandgap. Qualitatively identical effects observed for other self-assembled quantum dots were associated with the thermally induced redistribution of carriers within the dot ensemble [8,10,22]. Thus, one may preliminarily conclude that, due to a strong interdiffusion activated by a high temperature of the growth, the fluctuations of e.g. chemical composition present in the structure are able to confine excitons. Then, the transfer of carriers between those fluctuations with a different depth of the corresponding potential causes the observed red-shift of the PL energy.

In order to analyze the temperature dependence of the PL intensity, which would provide the information about the non-radiative recombination in the structure, it is necessary to get rid of the influence of the carrier redistribution effects evidenced by the large red-shift of the energy. In other words, it is improper to trace the intensity change with the temperature just by following the shift of the respective energy peak of the PL emission obtained by fitting. Since, as it is going to be shown later, the energy shift with the temperature for excitons localized in a single QD is described by the temperature variation of CdTe bandgap energy, it is reasonable to apply the following procedure to determine the temperature dependence of the PL intensity. Namely, for the certain energy position in the spectrum measured at $T=4.2$ K the emission intensity as a function of the temperature was determined at the photon energy that follows the shift of the energy gap of CdTe. The energy gap dependence of CdTe on the temperature was assumed to be the same as reported previously for CdTe/ZnTe QWs [6] with the temperature coefficient equal to -0.34 meV/K. Such a procedure was carried out for several values of initial PL energy at $T=4.2$ K. The result of this analysis is presented in Fig. 3. The solid points are the intensities of the PL line measured for different energies marked by arrows in the inset of the figure. The line represents the PL intensity dependence on the temperature obtained for CdTe/ZnTe QWs [6]. For an easier comparison, each curve in Fig. 3 was normalized to its value at $T=4.2$ K. The obtained result

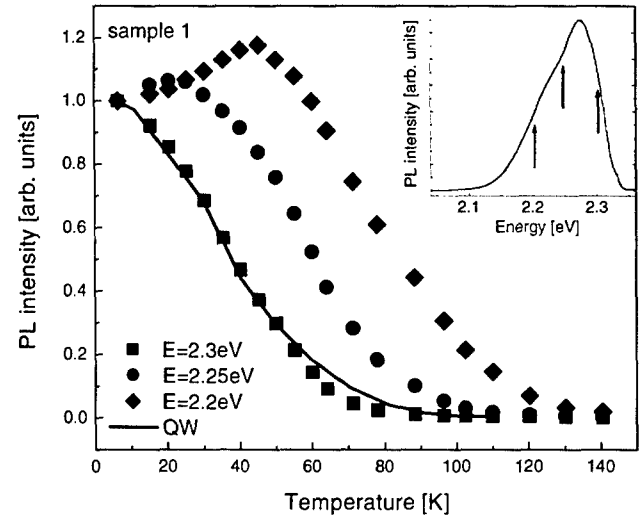

Fig. 3. The analysis of PL intensity dependence on the temperature performed for sample 1. The solid points are the intensities of the PL line measured for different energies marked by arrows in the inset of the figure. The line represents the PL intensity dependence on the temperature obtained for CdTe/ZnTe QWs [6]. For easier comparison, all data are normalized to the intensity at $T=4.2$ K.

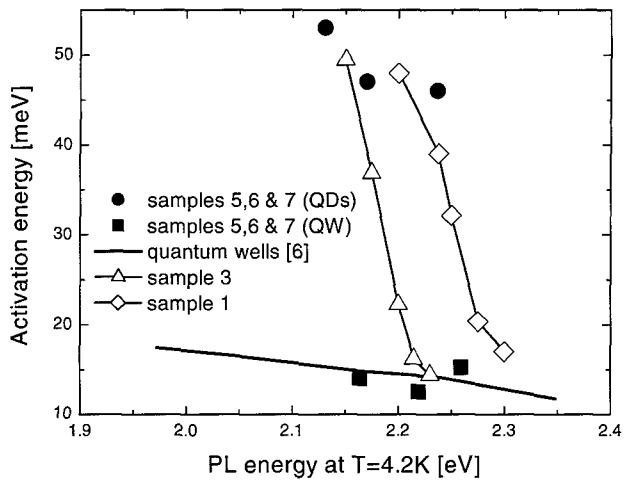

Fig. 4. The values of activation energies of the PL quenching as a function of the emission energy at $T=4.2$ K for different structures. The open points are the values obtained for samples number 1 (diamonds) and 3 (triangles). The squares and the circles represent the activation energies obtained for quantum well emission and QD PL measured for samples 5, 6 and 7. The solid line gives the values reported previously for CdTe QWs.

reveals a strong dependence of the PL intensity temperature behavior on the energy. For the highest energy, namely 2.3 eV (squares in Fig. 3), the intensity dependence is identical as the one obtained for a pseudomorphic QW (solid line). This indicates that the recombination at this energy is predominantly related to excitons localized in a QW-like potential. Recombination of these excitons quenches rapidly with increasing temperature due to their dissociation. This may explain fast disappearance of the high-energy peak, as depicted in Fig. 2. In contrast, the low-energy band is observed at much higher temperatures. This is clearly evidenced in Fig. 3, where the diamonds represent the temperature behavior of the intensity of this band. Up to 50–70 K the emission remains constant or its intensity even slightly increases. Such an effect is commonly recognized as a characteristic QD behavior [8,9,23].

Some more quantitative information about the excitonic localization in the quantum structures can be obtained by estimating the activation energies of the PL thermal quenching. To fit the data the following formula was used:

$$I = I_0/[1 + C\exp(-E_A/kT)]$$

where E_A is the thermal activation energy of the excitonic emission, C is the ratio of non-radiative to radiative transition probabilities and I_0 is the proportionality constant. The results are presented in Fig. 4, where the activation energy of the PL quenching as a function of the emission energy at $T=4.2$ K is plotted.

The open points represent activation energy obtained for different emission energies for samples 1 (diamonds) and 3 (triangles). Additionally, the results obtained for samples 5, 6 and 7 are shown: the squares and the circles represent the activation energy evaluated, respectively, for the two-dimensional QW emission and for QD-related luminescence. The solid line gives the values reported previously for pseudomorphic CdTe/ZnTe QWs [6].

The comparison between the activation energy obtained for samples grown at different conditions indicates that in the case of structures grown either at $T=$ 480 °C or with a long healing after CdTe deposition a gradual evolution in the exciton confinement takes place. Namely, starting from two-dimensional QW-like localization, with a decrease of the emission energy, the excitons become further localized in all three directions, resembling that in QD. Thus, we conclude that the enhancement of the atom mobility during the growth leads to the formation of quantum dot-like objects. This conclusion is additionally corroborated by the presence of clearly separated emission bands related to QW and QD emissions in structures where both methods were employed during the growth: the high substrate temperature and the healing after QD layer deposition.

The results of PL temperature dependence which allowed us to distinguish between QW-like and QD-like exciton localization let us also concentrate in the following only on one type of the exciton confinement, namely that of QD zero-dimensional excitons.

3.3. Time-resolved photoluminescence

The degree of the excitonic localization influences not only the steady state optical properties but has also its strong impact on the dynamics of excitons in quantum structures. As an example of the typical behavior, the transients collected at $T=1.8$ K for several detection energies for sample 1 are presented in Fig. 5. In the inset, the extracted values of the decay time are also shown together with the PL spectrum. A gradual increase of the decay time value from 140 to 400 ps with a decrease of the detection energy was found, similarly to other systems with a large scale of potential fluctuations [24,25]. This increase, however, is not purely related to a possible dependence of the decay time on the size of the quantum dot but rather is an evidence of some exciton scattering mechanism present in a QD ensemble. One probable explanation of the observed behavior would be the phonon-assisted tunneling between different quantum dots. Since the results of Fig. 5 were obtained at low temperature one can exclude the optical phonon contribution in this kind of process. Thus, just low energy acoustic phonons could take part in such a mechanism. In particular, the exciton–acoustic phonon scattering processes were found to be responsible for the similar effects in thin CdTe/ZnTe QWs [26].

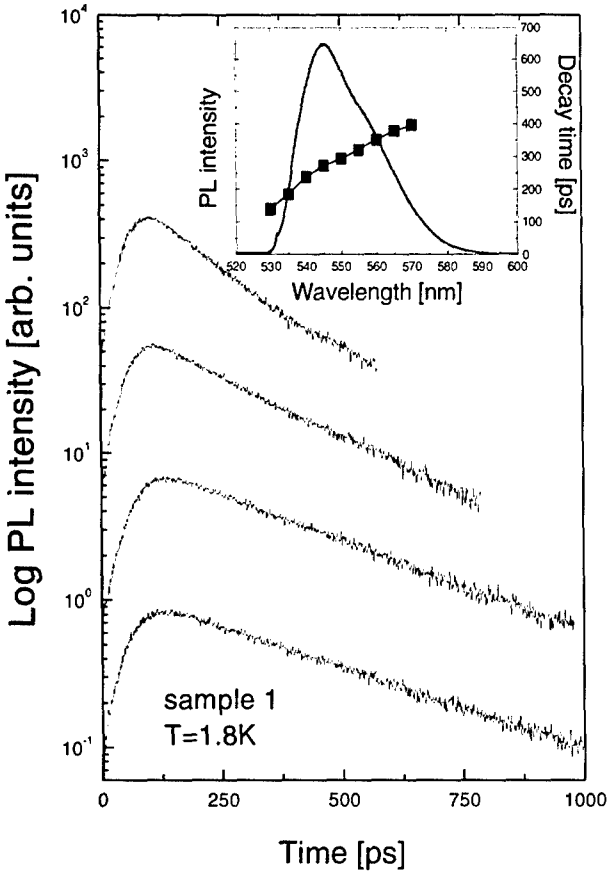

Fig. 5. The transients collected at $T=1.8$ K for several detection energies for sample 1. In the inset, the extracted values of the decay time are shown together with the PL spectrum.

Closer inspection of the time-resolved spectra reveals, apart from the main decay discussed above, also a second component, which is of approximately two orders of magnitude lower in its intensity. Moreover, it is substantially longer decaying, reaching up to 9 ns. When studying the temperature behavior of the time-resolved PL for all structures collected in Table 1 it is found out that, as shown in Fig. 6, the intensity of the long time component decreases quite fast with increasing temperature. In contrast, the intensity connected to the faster decay stays unchanged up to even $T=100$ K. Furthermore, the value of the shorter decay constant is independent of the temperature until the redistribution of carriers between the quantum dots with different localization potentials starts to influence the optical properties. Namely, for temperatures higher than 30 K, the decay time starts to depend on the excitation power. These effects are going to be discussed elsewhere [27].

The presence of two decay times was very recently reported for QDs in other material systems, in particular for CdSe QDs in ZnSe matrix [15]. The origin of this decay is still under debate, the possible explanations will be considered in the next part of this paper devoted

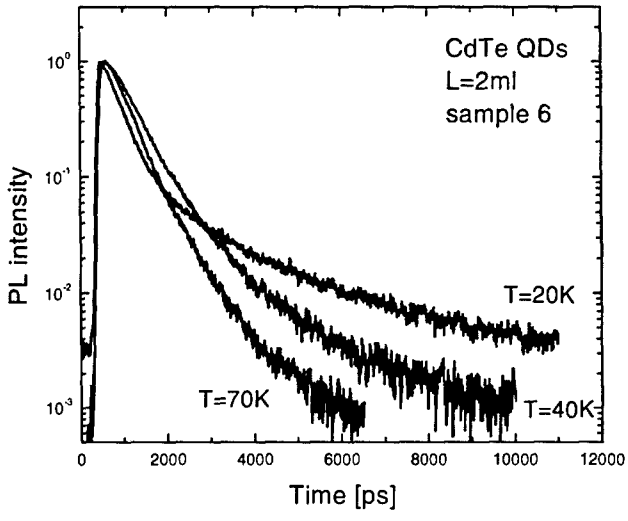

Fig. 6. Temperature behavior of the time-resolved PL obtained for sample 6. The transients are normalized to the maximum intensity.

to the temperature dependence of the microluminescence of QDs.

3.4. Microluminescence as a function of the temperature

When decreasing the number of the quantum dots accessed in optical experiment, for example by reducing the size of the laser spot, the broadened spectrum splits into the sharp spikes related to recombination in single dots. This is in contrast to the case of QW, where for free excitons the PL emission does not depend on the size of the probed area of the structure. In Fig. 7 the results of a microluminescence at $T=6$ K obtained for the sample with 10 layers of CdTe QDs with a nominal thickness equal to 2 ML are presented. According to the expectations, the spectrum measured with a micrometer size laser spot consists of a series of narrow emission lines superimposed on a broad PL band. The line pattern

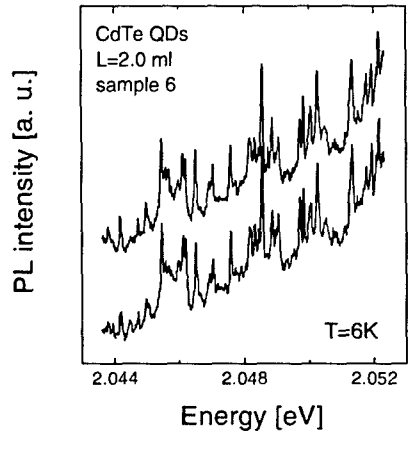

Fig. 7. Microluminescence results obtained at $T=6$ K for sample 8. The two spectra were subsequently one after another.

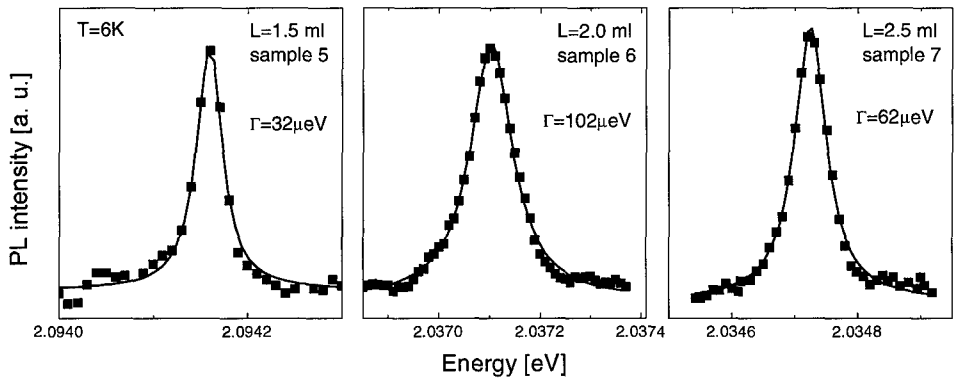

Fig. 8. The comparison between individual QD emission lines measured experimentally and fitted by the Lorentzian shape. The spectra were obtained for samples 5, 6 and 7.

is reproducible, as evidenced in Fig. 7, where two spectra taken subsequently one after another are shown. It is clear that the observed line sequence changes when the laser spot moves from one place on the sample to another. This fact demonstrates, indeed, that the quantum dots with different parameters such as size and/or chemical composition are participating in the total ensemble broad emission shown in Fig. 1c.

The linewidth of the individual lines was estimated by fitting a Lorentzian shape to a spectrum of the single dot and it was found to be in the order of 100 μeV. In Fig. 8 the comparison between experimentally obtained lines ($T=6$ K) and those fitted is given for three samples with different nominal thickness of CdTe layer. The Lorentzian shape of the line is due to the fact that an ideal quantum dot one should consider, in principle at least, as a homogenous system (no inhomogeneous broadening). There were, however, recent reports showing that due to the interaction between a QD and its neighborhood, the PL emission associated with that particular dot could exhibit fluctuations of both its energy and its intensity [28,29]. Also internal degrees of freedom such as spin in magnetic QDs can contribute to modification of the linewidth of the PL emission [30]. The effect reported in [28,29] was, however, ascribed to the changing of the charge configuration in a QD surrounding induced for example by structural defects. If such an effect is present then, obviously, the measured linewidth strongly depends on the relation between the duration time of the spectra acquisition and the characteristic time of charge configuration changes in QD surroundings. If the latter is much shorter, then the measured linewidth will considerably increase with the acquisition time. In an opposite case, the linewidth measured for a single dot should not depend on the time of collection of the spectra. In the case of CdTe QDs studied in this paper the width of the lines related to a single QD emission remained unchanged within the timescale of hundreds of second. Additionally, we did not observe any influence of either excitation power or acquisition time on the single dot PL energy and on the emission line width. The absence of such effects indicate that our structures are either free from defects (which was further confirmed by transmission electron microscopy measurements, where no signature of dislocations or other structural imperfections were observed) or that the charges on those defects that exist are frozen within the timescale of collecting the spectra (typically approx. 1 min).

The temperature evolutions of the microluminescence spectra obtained for the samples number 5 and 6 are presented in Fig. 9. The spectra in Fig. 9a are normalized in order to allow an easy comparison. The main result of these experiments is that a sharp structure composed of the lines related to single QD emissions quenches with an increase in temperature, while the broad background survives up to much higher temperatures, similarly to the case of CdSe QDs, as recently observed [15,16]. Smearing out of the sharp lines correlates with a decrease of the PL intensity related to the long-decay component in the time-resolved spectra, as described earlier (Fig. 6). This correlation allows, then, to assume that both behaviors are caused by the same effect. Such an assumption is supported by recent experiments performed on a single dot, which reveals the presence of two decay times in the time-resolved PL spectra [15], with exactly the same values as those obtained by means of macro-PL measurements. On the basis of these results we can ascribe the spread of the microluminescence spectrum shown in Fig. 9a to an exciton–acoustic phonon scattering in QDs. This scattering process was found to be significantly enhanced in QDs as compared with excitons confined in systems of higher dimensionality. Therefore, it can give rise to the broadening of the line with temperature [17].

By following the single QD-related emission line it is possible to extract the temperature dependence of its energy shift and its linewidth, as presented in Fig. 9b. The energy of a single emission line was found to follow the shrinkage of the bandgap of CdTe (solid line

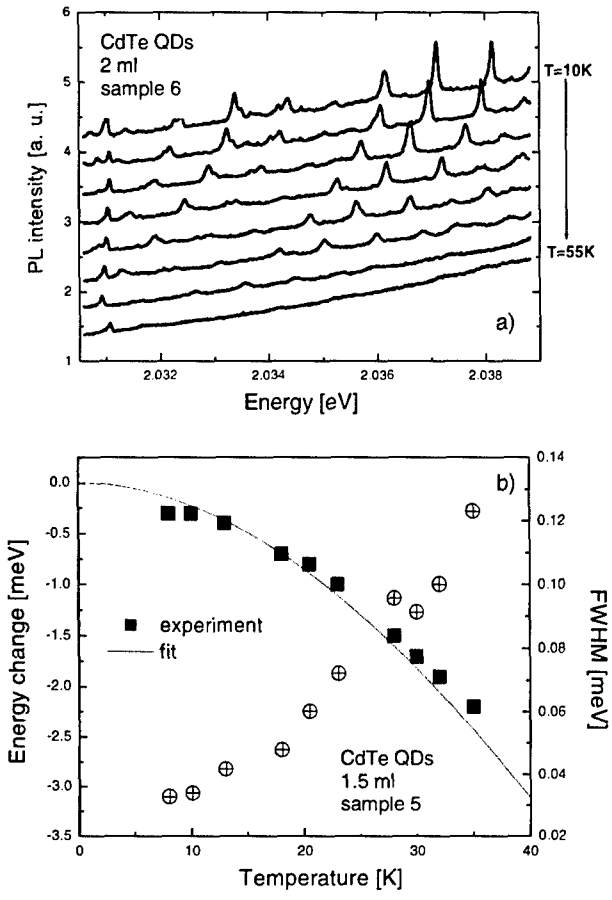

Fig. 9. (a) The temperature evolution of the PL spectra obtained for sample 6. The spectra are normalized for an easier comparison. (b) typical energy and linewidth dependence of the single exciton line obtained for sample 5. The solid line represents the CdTe bandgap temperature shrinkage, while points are experimental data.

in Fig. 9b) for most of the dots investigated. However, the linewidth increases superlinearly with the temperature as reported for GaAs QDs [13]. Contrary to this behavior, in both bulk semiconductor material and in QWs, the homogenous linewidth of the excitons increases linearly [13]. As discussed in [13] this is a direct result of the δ-like density of states in QDs.

The single QD spectroscopy provides also the insight into a particular in-plane symmetry of the dot. From theoretical considerations [31,32] it is known how the exchange electron-hole interaction and the symmetry act on the internal exciton level structure. In particular, for the in-plane symmetric QD (with a circular in-plane potential) the bright exciton level ($m=\pm 1$, m representing the effective spin of exciton) is two-fold degenerate and it is split from the dark exciton states ($m=2$ and $m=-2$) by the exchange interaction. However, if the dot is elongated (asymmetric) which means that the symmetry is lowered, the bright level degeneracy if lifted.

In Fig. 10 the spectra measured for three different quantum dots are shown in two linear polarizations, along (110) and (1−10) crystallographic directions. It is evidenced that both the sign as well as the magnitude of the splitting vary from one dot to another. Moreover, when analyzing the splitting for larger number of dots it was found that these two parameters were distributed randomly within the ensemble, with no systematic trends. Since the splitting between the two linearly polarized components is caused by the in-plane anisotropy of the dot potential [9,13,16,18], then from the obtained results we may conclude that there is no preferential orientation of the elongation of the QDs in our structures. The results of the PL measurements performed on the entire QD ensemble, where no sign of the anisotropy of linear polarization was found, support additionally this conclusion.

3.5. Microluminescence as a function of magnetic field

Single QD spectroscopy performed as a function of magnetic field offers a possibility to study the influence of the confinement and the symmetry on the diamagnetic shift and on the Zeeman splitting of excitons [3,11,18]. In Fig. 11 the PL spectra for two dots with different in-plane symmetry are presented as functions of the magnetic field. The data were obtained for sample 9 through apertures in a sputtered metal mask. In both cases with an increase of the magnetic field the splitting between the lines observed in different circular polarizations

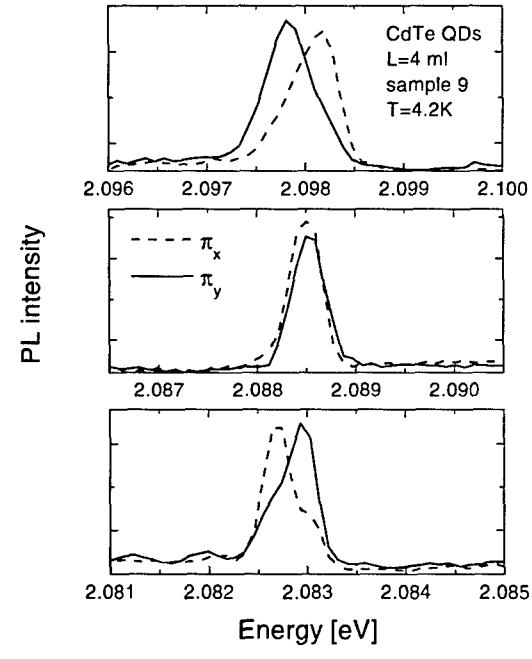

Fig. 10. Zero-field PL spectra measured for three different quantum dots in two linear polarizations. The results were obtained for sample 9.

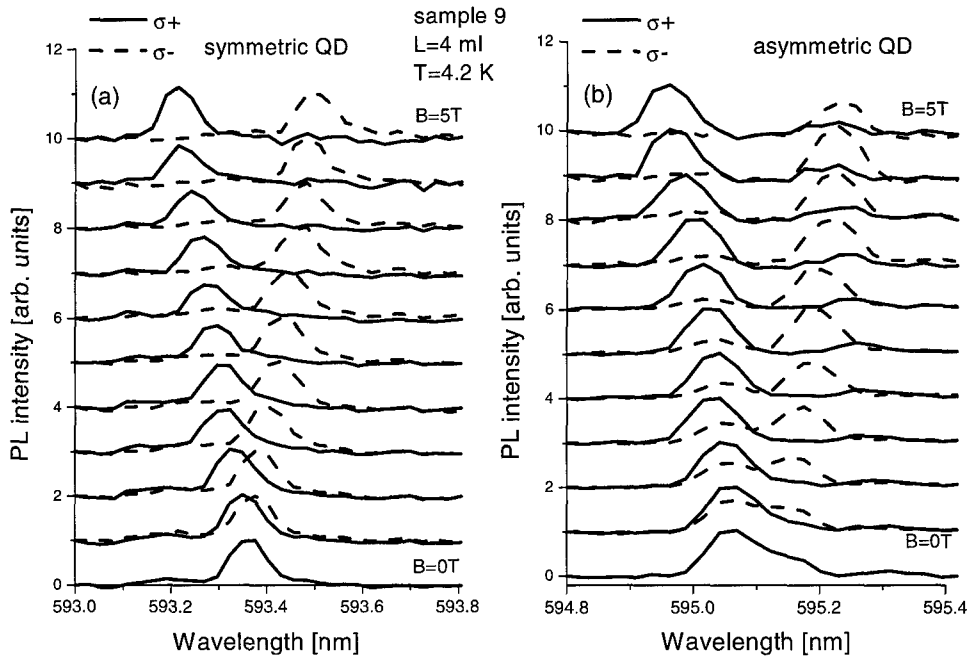

Fig. 11. PL spectra obtained for the sample 9 at $T=4.2$ K showing the magnetic field behavior of two QDs with different in-plane symmetry.

increases. Moreover, both spin-up and spin-down emission lines are slightly shifted to high energy due to the diamagnetic shift of the exciton line. The fact that the two emission lines are observed for every single dot indicates that the spin relaxation time of excitons in our CdTe QD is much longer that the time of the radiative recombination. Indeed, as shown in [33], the linear polarization of the QD ensemble emission does not change at the time scale of several tens of nanoseconds, although the decay time of excitonic recombination is equal to 700 ps.

The influence of the in-plane symmetry on the optical properties is observed in the polarization behavior of the emission. Namely, in the case of the symmetric dot (left panel) the emission is almost completely circularly polarized already at $B=0.5$ T. Contrary to that, the asymmetric QD emission is not completely polarized, and even at the highest magnetic field used ($B=5$ T) the degree of the circular polarization does not reach 100%. This is a consequence of the initial linear polarization of the PL doublet related to in-plane asymmetry of the zero-dimensional potential.

Concerning the exchange energy in CdTe QDs studied in this work the only solid information, which appears from the experimental results, is that the value of the splitting between the bright and the dark excitonic levels is larger than the Zeeman splitting caused by the magnetic field. If it were the opposite case, then it would be possible to observe the dark levels due to the mixing with the bright states, as reported for larger quantum dots [3]. Since, obviously, the magnitude of the exchange interaction increases with a decreasing size of the dot [34], then the absence of the levels characterized by $m=\pm 2$ is not surprising for such small quantum dots as studied in present paper.

The model presented in Kuther et al. [11] and Kulakovskii et al. [18] enables to evaluate the extension of the exciton wavefunction in a QD taking into account the diamagnetic shift of the exciton. The value estimated for our structures is equal to 3 nm, which agrees reasonably with the size of the dots obtained from transmission electron microscopy investigations [1]. Furthermore, by fitting the magnetic field dependence of the energy, the value of effective exciton g-factor was obtained to be equal to -3. The value is almost twice that obtained in bulk CdTe indicating that the g-factor increases with a degree of quantum confinement. By analyzing the diamagnetic shift and the value of the exciton g-factor for several tens of QDs it was found that both these parameters exhibit no systematic dependence on the zero-field PL energy. This insensitivity to the ground state QD energy may suggest that the confinement of excitons is quite homogeneous within the QD ensemble. The assumption is justified to some extent by the result of theoretical modeling of CdTe QDs [35] showing that in order to reproduce the PL emission shown in Fig. 1d the size should vary from 2.6 to 3.4 nm. Thus, with respect to the CdTe exciton Bohr radius (10 nm in bulk) this change is negligible.

4. Conclusions

The optical properties of CdTe/ZnTe quantum structures in dependence of their growth conditions were

discussed. It was shown that growth either at a high temperature of 480 °C or subjecting the CdTe layer to a healing after its deposition lead to formation of quantum dots. The self-assembly of quantum dots is also achievable when a combination of these two techniques as well as applying atomic layer epitaxy when growing CdTe at 320 °C are used. Optical spectroscopy allows unambiguous distinguishing of the type of the excitonic confinement present in a particular structure. The QD emission is observable up to temperatures of approximately 200 K, considerably higher than in the case of CdTe/ZnTe QW structures. Moreover, the QD PL is characterized by two decay times at low temperature: one at approximately 300 ps and the other one, which is more than the order of magnitude longer. When decreasing the laser spot size, the QD spectrum splits into a series of sharp lines related to exciton recombination in single dots. The temperature dependence of the long decay time as well as the disappearance of the sharp lines with increasing temperature is related to exciton–acoustic phonon scattering in single dots. The single dot spectroscopy reveals that some of the dots have an oblong in-plane shape, however, no preferential direction of this elongation is found. From the magnetic field dependence of the single dot PL the values of diamagnetic shift and effective exciton g-factor are estimated. Both these quantities show are independent on the ground state emission energy of the dot as well as of its particular in-plane symmetry.

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